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Title:	Atomistic Modeling of He in Fe Using *ab initio* and Semi-Empirical Potentials		
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Atomistic Modeling of He in Fe Using *ab initio*

and Semi-Empirical Potentials

S.G. Srivilliputhur, M.R. James, M.I. Baskes, and S. Maloy

1. Introduction

It is well known that materials undergo radiation damage from the impingement of neutrons and protons emitted nuclear environments such as fission reactors, fusion reactors and accelerators. While there have been many successful efforts in modeling this process, modeling of the interactions and the damage induced by H and He, the main gaseous by-products of the nuclear reactions, on the underlying materials (usually ferritic steels) is still unclear. Because of elevated H and He production from damage due to high-energy irradiation, one needs to urgently expand materials' properties database. A major problem is the limited amount of reliable experimental data. There is a paucity of even the basic experimental data on defect formation and migration energetics and phase stability of H-Fe/Fe-He systems that are of interest to this project. It is important to realize that the laboratory experiments (irradiations) are often cost-prohibitive.

With the advancement of computer technology, modeling and simulation have emerged as an indispensable tool to bridge this gap in our experimental understanding. Theoretical approaches based on state-of-the-art *ab initio* electronic structure calculations are therefore most desirable. However, these calculations are computationally intensive and allow only a few hundreds of atoms in a system. On the other hand, semi-empirical many-body interatomic potentials such as the modified embedded atom method (MEAM), based on the density functional theory, allow us to handle large systems with millions of atoms [1]. This is a powerful enhancement of the classical embedded atom method (EAM) and allows us to handle directional forces, important in ferritic materials [2]. Furthermore, in order to extend a potential to model alloys such as He inclusions in ferritic steels, it would be convenient to describe the atomic interactions of the multitude of alloying elements using a common formalism.

Thus, for our modeling work we will develop and use interatomic interactions based on the MEAM model.

We succeeded in our ultimate goal for the year 2003: *develop a reliable MEAM model for Fe-He system in order to understand the effect of He on the properties of Fe.* Using this potential we have investigated the phase stability, defect energetics, defect mobility, and mechanical properties of the Fe-He system using a combination of molecular dynamics and thermochemical calculations. The ultimate goal is to use MEAM to investigate effect of radiation dose, defect microstructure, and gas inclusions on macroscopic materials properties.

To create a MEAM model for Fe-He, we first need a sufficiently large database of properties of the Fe-He system. Since reliable experimental values of properties are lacking, we first used *ab initio* calculations to generate a database of many equilibrium properties. This dataset will then be used to determine a reliable MEAM model of He-Fe.

2. First Principles Modeling of Fe-He

To develop the MEAM model we first generated a database of various equilibrium properties of Fe, He, and Fe-He systems using state-of-the-art *ab initio* electronic structure calculations. These properties are shown in Figure 1 and summarized in Tables 1 and 2. These properties in conjunction with known experimental properties of Fe were used to develop a MEAM model. Currently, we are refining the model.

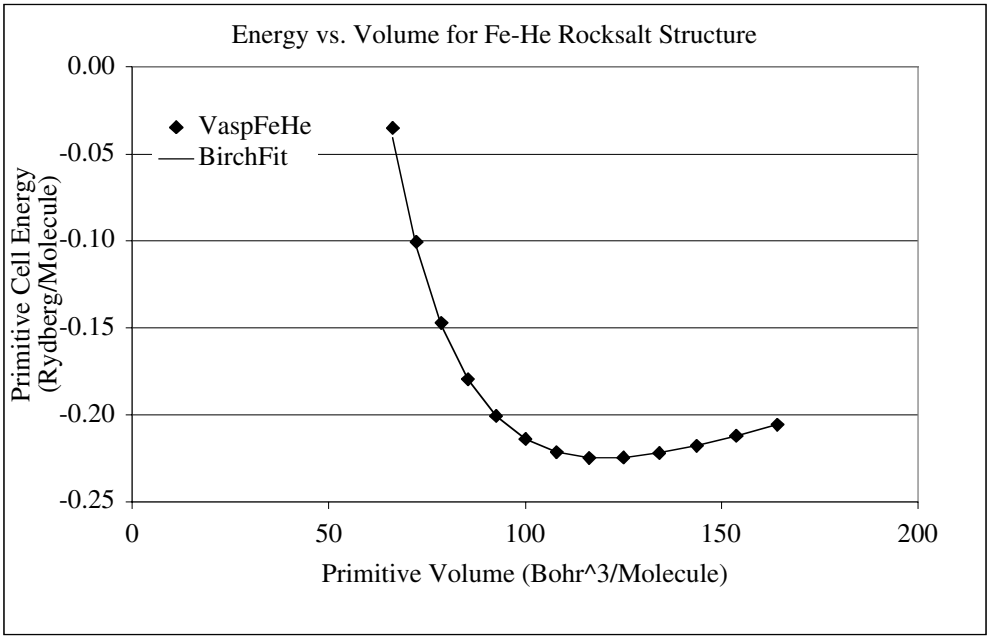

Figure 1. *Ab initio* calculations of Fe-He rocksalt structure. These calculations give us the equilibrium volume and cohesive energies and are used to develop MEAM model.

Table 1. Properties of Fe-He rocksalt structure computed using *ab initio* calculations.

Property	BCC
Cohesive Energy	3.06 eV
Lattice Constant	4.14 Angstroms
Bulk Modulus	66.3 GPa
C_{44}	-20.2 GPa
$(C_{11}-C_{12})/2$	-108.4 GPa

Table 2. Properties of FCC He structure computed using *ab initio* calculations.

Property	BCC
Cohesive Energy	0.032 eV
Lattice Constant	4.1 Angstroms
Bulk Modulus	1.57 GPa

3. MEAM Interaction Model for Fe-He

Next, we developed a MEAM interaction model for the Fe-He system using data from both experiments and first principles calculations. Properties such as cohesive energies, elastic constants, equilibrium lattice constants, and vacancy formation energies of the body centered cubic and face centered cubic Fe were used for fitting pure Fe MEAM model. For He interaction, we used equilibrium lattice constants, elastic constants, and cohesive energies from first principles and experiments. For Fe-He interaction, we used cohesive energy, elastic and lattice constants of an ideal Fe-He rock salt crystal obtained from first principles. This MEAM model reproduces the properties in the fitting database well. It also *predicts* many properties, which are known experimentally, but were not used for fitting the potential quite well. Lastly, we used the Fe-He MEAM model to predict many point defect properties that are not available experimentally. The computed properties look very reasonable and give us confidence in the developed MEAM model. This model was presented in the 2003 AccApp Meeting in San Diego. Some of the predicted properties of Fe (not used for fitting) are shown in Figure 2 as a histogram. Figure 3 plots point defect properties of pure Fe, and Figure 4 shown the formation energies of He interstitials in α-Fe.

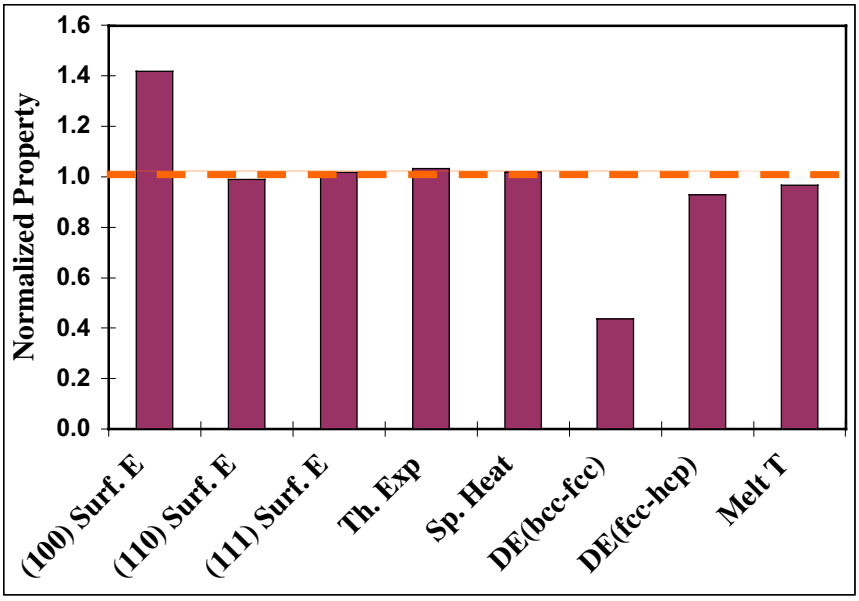

Figure 2. Histogram of some properties predicted for pure bcc Fe (not used in fitting database). These properties were normalized by their corresponding experimental values. If there is good agreement between MEAM and experimental properties, the normalized value will be 1 (shown by dashed line). The (100) surface energy ((100) Surf. E.) looks unreasonable because the experimental value of surface energy of a polycrystal was used for normalizing and it contained very little (100) surfaces.

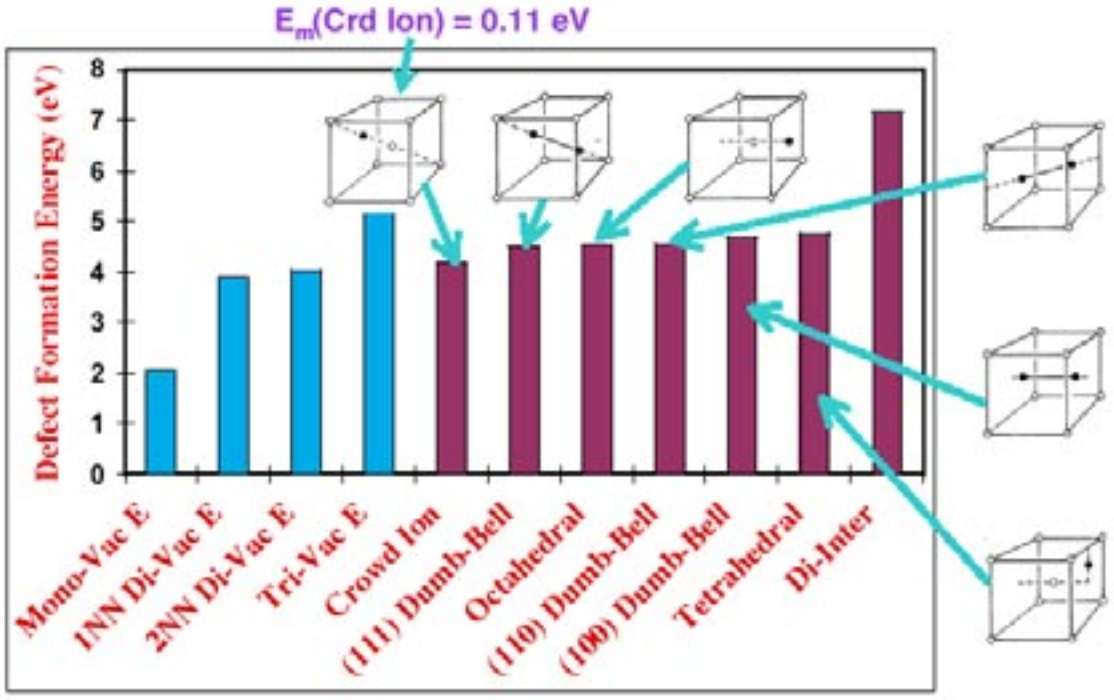

Figure 3. MEAM Fe yields reasonable point defect properties for pure bcc Fe.

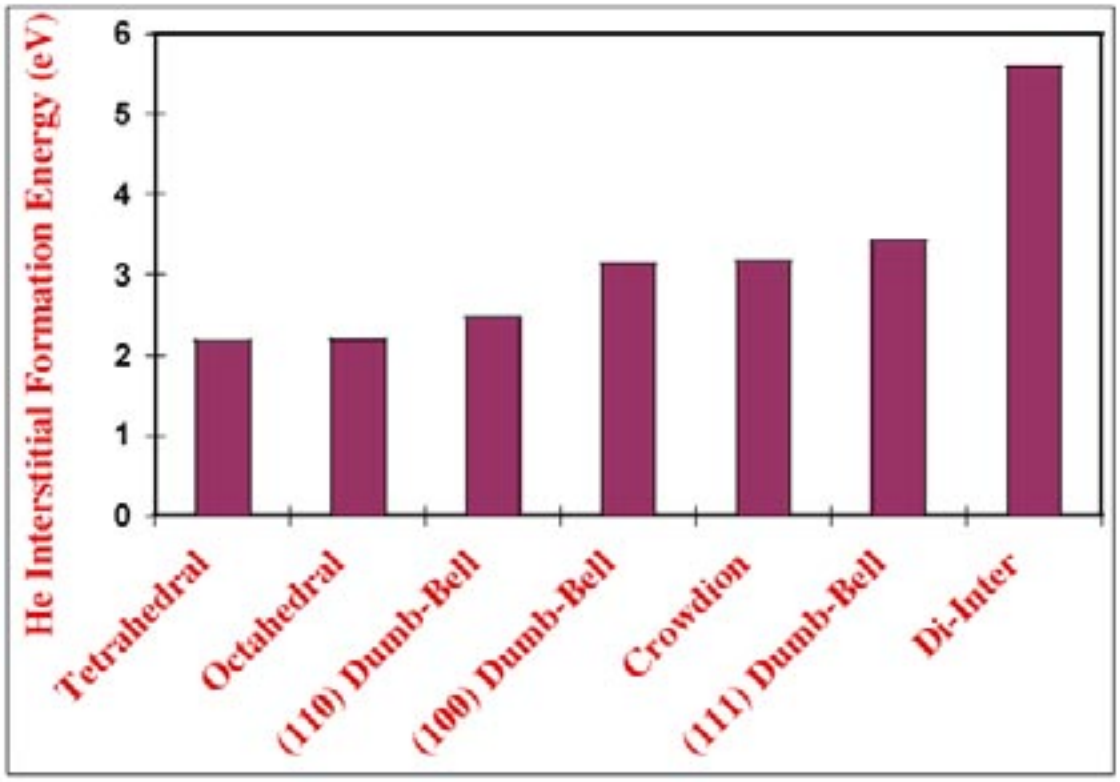

Figure 4. He interstitial formation energy in α-Fe. MEAM predicts stable He interstitial clusters with up to 5 atoms in the absence of vacancy and up to 9 atoms when there is one vacancy at the Fe site.

In summary, the developed MEAM model gives reasonable values for various defect properties of the Fe-He system.

4. References

[1] M.I. Baskes, Phys. Rev. B. **46**, 2727 (1992); B-J. Lee and M.I. Baskes, Phys. Rev. B **62**, 8564 (2000).

[2] M.I. Baskes, Phys. Rev. B, **62**, 15532 (2000).